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ABSTRACT Over the past two decades, we observed a substantial rise in ionic content that was mainly determined by the sulfate concentration at 20 remote high elevation lakes located in central southern Himalaya. At LCN9, which was monitored on an annual basis for the last twenty years, the sulfate concentrations increased over 4-fold. Among the main causes, we exclude a change in the composition of wet atmospheric deposition, as well as a possible influence of decrease in seasonal snow cover duration, which could have exposed larger basin surfaces to alteration processes. Glacier retreat likely was the main factor responsible for the observed increase of sulfate concentrations. We attribute this chemical changes mainly to the sulfide oxidation processes that occur in subglacial environments. Moreover, we observe that the weakened monsoon of the past two decades has only partially contributed to the lakes enrichment through runoff waters that are more concentrated in solutes or lowering the water-table, resulting in more rock exposed to air and enhanced mineral oxidation.

INTRODUCTION

High mountain ecosystems are especially vulnerable to climate change, and lakes in particular may act as sentinels.[1,2] An evaluable opportunity for a fine-scale investigation is particularly evident on the south slope of Mt. Everest (Nepal), which is one of the most heavily glacierized parts of Himalaya[3]. At the same time, this region is the most densely populated by glacial lakes in the overall Hindu-Kush-Himalaya range[4]. In this region, Lami et al.[5] observed a significant relationship between the increase in the annual temperature recorded in the area and the enhanced conductivity detected in two glacial lakes located above 5000 m a.s.l. in the last decades, but without inferring any undergoing physical process or source of ionic release. At the same time, an increase in conductivity and sulfate concentrations has been

observed in a few lakes of the European Alps, but in this case, the changes have been specifically ascribed to the solute release from an active rock glacier as a response to climate warming[6]. An increase in sulfate from melting ice during drought years is also reported by Williams et al.[7] in the outflow of a rock glacier in the Colorado Front Range and by Mast et al.[8] in southwestern Colorado. Even in this last case the authors consider temperature as the main climatic driver of change and sulfate as largely derived from pyrite (watershed weathering sources), although the undergoing physical processes are only supposed.

In this study, after exploiting the recently reconstructed[9] daily temperature and precipitation time series and the available accurate tracing of the glacier shrinkage in the region[10], we discuss changes in the water chemistry of 20 remote lakes located on the south slopes of central Himalaya that were sampled approximately after twenty years, starting in the early 1990s. Furthermore, during this period, two selected lakes (named Lake Cadastre Number (LCN)[11] 9 and 10) were monitored yearly. We consider several drivers of changes of the in-lake chemistry, including temperature, precipitation, atmospheric deposition, rocks and soil weathering processes (periglacial environment), seasonal snow cover duration, and, in particular, the role of glaciers (subglacial and surrounding periglacial components). With regard to permafrost, little is known about its distribution in Himalaya.[12] Therefore the subsurface ice and its possible degradation is not included in this analysis, although its influence on in-lake chemistry evolution cannot be excluded as reported by other authors[13,14] in the Colorado Front Range.

MATERIAL AND METHODS

Study area. The monitored glacial lakes are located in Sagarmatha (Mt. Everest) National Park (SNP), Nepal, which lies in the central southern Himalayas (Fig. 1a, 1c). The SNP (1148 km^2), crossed by the Khumbu valley, is the world's highest protected area, extending from an elevation of 2845 to 8848 m a.s.l..[15,16] According to Searle et al.[17], the Khumbu valley substratum is dominantly made up of crystalline rocks (e.g,, sillimanite gneisses calc-silicates, amphibolites and K-feldspar augen gneisses) and leucogranites that intrude the metasediments as dykes, sills and bodies. The overlying Everest series (weakly metamorphosed shales and pelites with limestone bands), the Yellow band unit (limestones, marbles and calc-silicates) and the Ordovician limestones are only present in the eastern part of the basin. Extensive moraine debris of different ages, alluvial deposits and glacio-fluvial deposits host sub-surface aquifers in close connection with the hydrographic network.

The land cover classification shows that almost one-third of the territory is characterized by temperate debris-covered glaciers, with 75% of glacier surface area lying between 5000 m and 6500 m a.s.l.[10] and with less than 10% of the park area being forested[15,16]. The soils in the high valleys are primarily Entisols.[18]

Salerno et al.[19] reported the presence of 170 glacial lakes and provided an accurate description of their topographic characteristics. Most of them are small (median surface of 0.01 km^2) and within an elevation zone ranging from 4800 to 5300 m a.s.l.. Table 1 shows the main morphometric features of the 20 survey lakes. They have been selected to cover wide elevation (4466-5427 m a.s.l.) (Fig. 1b), basin slope (9°-35°), and basin aspect (120°-231°) ranges. The mean glacial coverage is 18%, with three basins without glaciers (LCN40, LCN66, and LCN70). Two sub-basins (LCN10 and LCN67) does not have glaciers within the sub-basin, which directly drain into the lakes, while in this case their upstream lake basin (LCN9 and LCN68, respectively) presents a glacier coverage (all data are reported in Table SI1). Details on these lakes and their basins can be found in Tartari et al.[20]. All glaciers lying within the survey lake basins are debris-free and without clearly detectable accumulation and ablation zones.

The climate is characterized by the monsoons, with a prevailing S-N direction.[21] During the last twenty years at the Pyramid meteorological station (5050 m a.s.l., Fig. 1c)[9], which is located just 500 m from LCN9 (Fig. 1c), 90% of the precipitation is concentrated during June-September, and the probability of snowfall during these months is very low (4%), whereas the amount of snowfall at an annual level reaches 20%. Snow as a percentage of total precipitation increases with the elevation gradient justifying the wide glacier coverage in SNP. Recently, the daily temperature and precipitation time series of the last twenty years (1994-2013) were reconstructed by Salerno et al.[9] for these altitudes (Fig. 1b), which correspond to the median elevation of the 20 survey lakes. The total annual cumulated precipitation at this elevation is 446 mm, with a mean annual temperature of -2.45 °C. The mean annual air temperature has increased by 0.9 °C since the early 1990s. The significant increases were experienced just for the months of April, November, and December.[9] As the air temperature increased, the average elevation of the freezing line (0 °C isotherm) has moved upward. The major changes occurred in April, where it moved upward by 225 m, passing from around the mean elevation of glaciers terminuses to the mean elevation of the surfaces of the same glaciers located in the basins of the 20 survey lakes (Fig. 1c). The increase in air temperature observed at these high elevations during April fits with the warming reported by Pal and Al-Tabbaa[22], who observed that only April shows significant changes in all Indian regions and the West Himalaya (1901–2003 period). As for precipitation, a substantial reduction of rainfall (47%) and in the probability of snowfall (-10%) has been observed in this area over the last twenty years[9]. . According to Yao et al.[23] there is strong evidence of the general monsoon weakening even in the overall Himalayas.

Water chemistry analysis. The chemical composition of 20 remote lakes located in SNP was evaluated in the period between 1990 and 1997 (hereafter early 1990s) and successively from 2008 to 2012 (hereafter early 2010s). Lake sampling was carried out in autumn, at the end of the monsoon season. Samples were collected from the lake shore, possibly close to the lake outlet. Moreover, for the lakes LCN 9 and LCN10, sampling and chemical analyses have been performed annually since 1990 (1991 for LCN9), usually in late September or October, with the exception of the

missing years 1995-96 and 1998. For these lakes, three samples were generally collected along the water column from an inflatable boat in the deepest part of the lake. Here we took their average. For detailed information on the hydrochemical and biological characteristics of these lakes, see the work of Lami et al.[6].

All samples were collected using prewashed plastic bottles and stored at 4 °C for successive chemical analysis; these were performed during the 1990s by the CNR Water Research Institute and subsequently by the CNR Institute of Ecosystem Study (Italy). Several intercomparisons have been conducted to ensure data quality and comparability between the data produced by the two laboratories. Samples were analyzed for pH, conductivity, total alkalinity (using the Gran method for acidimetric titration), ammonium, total nitrogen, total phosphorus and reactive silica (by spectrophotometry), sulfate, nitrate, chloride, calcium, magnesium, sodium and potassium (by ion chromatography). Data were checked for internal consistency by means of the ionic balance and the comparison between measured and calculated conductivity. Details on the analytical methods and on the quality controls adopted in the laboratory can be found in Lami et al.[5].

In this paper, we focus on conductivity values, as an index of total solute concentrations of lakes, H^+, and major ions: calcium + magnesium (Ca^{2+}+Mg^{2+}), sulfate (SO_4^{2-}) and bicarbonate (HCO_3^-). These are indeed the prevailing ions in all sampled lakes, representing more than 80% of the total ionic content (Fig. SI1). We assume alkalinity as corresponding to bicarbonate ions, because at circum-neutral pH, as those of the study lakes, dissolved HCO_3^- is the dominant species contributing to alkalinity.

Morphometric and hydrological analysis. Seasonal snow cover duration of each survey lake basin has been attributed as a mean for the 2002-2012 period of the daily MODIS (Moderate Resolution Imaging Spectroradiometer) imagery (pixel 250 m) (http://modis.gsfc.nasa.gov/). The glacier surface areas within the survey lake basins were derived for the early 1990s from Landsat 5 TM (17 November 1992, pixel 30 m) and for the early 2010s from Landsat 11 ETM (30 November 2011, pixel 15 m) remote imagery. Data and methods are detailed in Thakuri et al.[10]. We interpreted the land cover of the remaining part of each basin through a visual interpretation of the Advanced Land Observation Satellite (ALOS, October 2008, pixel 10 m) remote imagery and distinguished terrain coverage by bare rocks, bare soils, and shrub vegetation. No rock glaciers were detected in these basins by visual interpretation.

Because we were interested in understanding whether the changes that occurred over the last twenty years in glacier surface areas and snow cover have influenced the solute variations observed during the same period, we had to overcome, as shown below, such limits linked with the resolution and availability of the satellite data.

Regarding glaciers, the satellite resolution of remote imagery available for covering the analyzed period provides a large uncertainty in estimating changes in surface areas, especially for small glaciers, such as those located within the basins of the survey lakes; thus, the estimated changes are dubious[19]. This limit has been overcome by enlarging the

scale of analysis to the overall SNP. At this scale, the 29 glaciers (with a surface area >1 km^2) that were already traced by Thakuri et al.[10] are more than 30 times larger (5.7±0.40 km^2 as median) than those located within the survey lakes (0.31±0.10 km^2 as median); thus, changes are more representative and less uncertain[19].

Concerning the snow cover, the MODIS imagery is available only since the early 2010s. For the previous decade, we deduced the trend in the coverage based on the estimated snowfall decrease reported in this area since the early 1990s[9]. The increase of temperature observed outside the monsoon period, when the precipitation is almost completely composed by snow, brought a significant decrease (-14%, $p < 0.05$) of the snowfall probability for the last twenty years at 5050 m a.s.l.. The trend reported above has been calculated by these authors according to Fujita and Sakai[24] and Ueno et al.[25] assuming that the probability of snowfall and rainfall depends on mean daily air temperature, using as thresholds 0 and 4 °C, respectively. The variations that occurred in the 2002-2012 period were computed as the difference between the first five and the last five years. Even in this case, the analysis was enlarged to the overall SNP to increase the representativeness within an elevation ranging from 4500 to 5500 m a.s.l. (6152 MODIS pixels), i.e., the altitudinal band of the survey lakes (Fig. 1b).

At this wide scale, we determined that changes in glacier surface areas and seasonal snow cover duration are related to certain morphometric boundary conditions. Elevation, aspect, and slope have been investigated for snow cover duration and glaciers, but for glaciers, we also considered the maximum, minimum and mean elevation, and the down and upstream slopes of the glaciers following the experience of Thakuri et al.[10] and Salerno et al.[19]. Detailed computational methods of these morphometric variables are reported in the works of these authors. In the same way, at the scale of each survey lake basin, we determined that the differences that occurred in SO_4^{2-} and HCO_3^- are related to the same morphometric boundary conditions investigated for glaciers and snow cover duration at a wider scale.

For simulating the daily melting of the glacier located in the LCN9 basin we used a T-index model[26]. This model uses the mean daily air temperature, the glacier elevation bands, and a melt factor (0.0087 m d^{-1} $°C^{-1}$) provided by Kayastha et al.[27] from a field study (Glacier AX010) located close to the SNP.

Statistical analysis. We conducted a Principal Component Analysis (PCA) among the lake chemical components and the basin morphometric features to obtain information on the relationships among the data and to look for reasons that could justify the observed changes in major ion concentrations[28]. The test was performed by using the "princomp" and the "biplot" functions in the R Project environment[29]. The degree of correlation among data was verified through the correlation coefficient (r) after testing that the quantile-quantile plot of model residuals follows a normal distribution. Otherwise data were log-transformed to meet the statistical requirements for normal distribution; then the residuals of the regressions were tested for homoscedasticity (not shown here) (e.g.,[29]). The chemical concentrations of the 20 survey lakes, measured in the early 1990s and the early 2010s, were compared, and the differences between the two

periods were tested by using the non-parametric ANOVA for paired comparisons (Friedman test) at $p<0.05$[28,30]. Differences in glacier surface areas and snow cover duration were analyzed by applying the same statistical test. The significance of annual chemical trends was evaluated with the annual Mann Kendall test at $p<0.05$.[31]

RESULTS AND DISCUSSION

Survey lakes. In the early 1990s, most of the survey lakes exhibited low values of conductivity (25 μS cm^{-1} as median) (Table 2, Fig. 2a). After approximately 20 years, the conductivity increased significantly, by 47% ($p<0.001$) on average. In some cases, the values even doubled. Lakes became significantly ($p<0.001$) enriched in solutes, primarily in SO_4^{2-} (+57%), then in Ca^{2+}+Mg^{2+} (+41%) and less in HCO_3^- (+11%) (Table 2, Fig. 2b,c,d). The increase in SO_4^{2-} was even 4-fold in LCN9 (from 84 to 327 μeq L^{-1}) and LCN10 (from 73 to 285 μeq L^{-1}) (Fig. 2b). For these lakes, a significant increasing annual trend ($p<0.05$) was observed both for conductivity, SO_4^{2-}, Ca^{2+}+Mg^{2+}, and HCO_3^- concentrations (Fig. 3b,c,d), with two main peaks in 1998 and 2010 (discussed in the last paragraph). All survey lakes (Table 2 and Fig. 2e), as well as LCN9 and LCN10 (Fig. 3e), exhibited a significant increase of hydrogen ion (H^+) concentrations (126%, $p<0.01$).

The temporal change of the relative contribution of the various ions to the total ionic content of LCN 9 can be observed in Figure SI1: sulfate passed from 22 to 36%, calcium remained stable (41%), while alkalinity contribution decreased from 24 to 11%. To assess the relative change in the ionic composition of the survey lakes, we calculated the c-ratio $HCO_3^-/(HCO_3^-+ SO_4^{2-})$, which is a geochemical indicator that is used to evaluate the relative dominance of the carbonation reaction and sulfide oxidation in meltwaters.[32] The c-ratio decreased significantly in the survey lakes (-18%, $p<0.01$) (Table 2, Fig. 2f). Particularly pronounced is the c-ratio decline observed in LCN9, from 0.56 to 0.29 (-48%), and in LCN10, from 0.59 to 0.31 (-47%) (Fig. 3f). The only exception with respect to the general pattern is LCN31 (Fig. 2f). The Duwo glacier, which is located within the LCN31 basin, is surging and shows a divergent response to climate change with respect to all of the other glaciers located in the region.[10] Therefore, we think that the two anomalies could be associated. All data presented in this section are reported in Table SI1.

In the following, we discuss potential factors that may have contributed to the increase of solute concentrations in the lakes, with special attention towards SO_4^{2-} and HCO_3^- ions: atmospheric deposition, weathering, precipitation, glaciation, and snow cover duration.

Atmospheric Deposition. The chemistry of wet deposition was investigated at the Pyramid station during 1991-1992[33] and 2007-2008[34]. The precipitation contents for all ions were the lowest ones among those measured at high elevation sites around the world[32]. The comparison between the studied periods revealed no substantial variations in SO_4^{2-} and other compounds (NO_3^- and NH_4^+) that resulted from anthropic activities, thus suggesting that the tropospheric

181 background concentrations are constant in the Himalayas[34]. These findings indicate that the atmospheric deposition
182 cannot play significant role in the solute increasing trend recorded in Himalayan lakes.

183 **Land cover and Snow cover duration.** We tested the possible dependence of SO_4^{2-} and HCO_3^- concentrations for the
184 survey lakes sampled in 1990s with respect to the basin land cover and seasonal snow cover duration. We preferred to
185 use 1990s data in order to investigate the dependences between these variables before a period (1990s-2010s)
186 influenced decidedly more by higher climate-driven changes[9]. The correlation matrix among all of these variables is
187 presented in Fig. SI3, and an overall overview of their mutual relationships is provided with the PCA in Fig. 4a. The
188 main finding is that the SO_4^{2-} concentrations are directly related to glacier coverage ($r=0.72$, $p<0.001$) (Fig. 4b), i.e.,
189 higher lake solute concentrations are found for basins with more glacial coverage. No relationship can be observed with
190 the other selected variables and in particular with the snow cover duration. Furthermore, we note that the HCO_3^-
191 concentrations do not have significant relationship with the land cover and the snow cover duration.

192 When observing the morphometric features of the LCN9 basin and the LCN10 sub-basin (Table 1), it becomes clear
193 that the discriminating feature between these lakes is the presence of a small glacier of 0.11 km^2 lying within the LCN9
194 basin, whereas the LCN10, which is located downstream, receives the glacial melting indirectly from LCN9 (Graphical
195 abstract). Concurrently, LCN10 has lower solute concentrations and conductivity (Fig. 3a, 3b, 3c, 3d). This fact can be
196 interpreted as further evidence that glaciers (subglacial and surrounding periglacial components) are the most important
197 driver of changes in both SO_4^{2-} concentrations and that the runoff in the non glaciated areas of the LCN10 sub-basin as
198 well as the possible groundwater discharge from headwater areas of the basin transports lower concentrations, thereby
199 diluting the LCN10 content. This interpretation is corroborated by the further evidence provided by lakes LCN68 and
200 LCN67. From Table SI1 we observe that the downstream lake (LCN67) has, among the other solutes, lower sulfate
201 concentrations (733 μeq L^{-1} respect to 926 μeq L^{-1} for LCN68). Even in this case the discriminating feature is the
202 presence of a glacier lying within the upstream lake basin (see Fig. SI2 and Salerno et al.[19]- Fig. SI2d-).

203 **Changes in glacier surface areas.** Thakuri et al.[10], considering glaciers larger than 1 km^2, reported a surface area loss
204 of -8.7%, $p<0.01$ for the overall SNP during the 1992-2011 period (uncertainty ±2.8%). Applying here the same remote
205 imagery and methods reported by these authors, but at glaciers located within the basins of the 20 survey lakes
206 (0.31±0.10 km^2 as median), we found a higher decrease in glacier area, but with an uncertainty connected with the
207 satellite resolution that increased by of an order of magnitude (-19.7%, $p<0.01$, uncertainty ±23.9%). The glacier within
208 the LCN9 basin lost 14% (uncertainty ±22%) of its surface area. According to many authors[10,35,36,37], even in SNP and
209 in the Himalayas in general, the main losses in area over the last decades have been observed for smaller glaciers
210 (glacier size vs glacier area change $r=0.58$, $p<0.01$, Fig. SI4). Therefore, the survey lake basins are subject to a glacier

shrinkage twice that observed for the glacial masses in the SNP region. However, the reduced size of these glaciers causes the area loss estimation to be affected by a large uncertainty that is even higher than its magnitude, and the estimation is thus too rough to be directly compared with the chemical variations observed in the relevant downstream lakes. Therefore, as described above, we conducted two parallel analyses: at the SNP level and at the scale of a survey lake basin.

At a larger scale, considering the data from Thakuri et al.[10], we analyzed which morphometric factors were able to cause changes in surface areas in the last twenty years (Fig. 5a). The correlation matrix among all of these variables is presented in Fig. SI4. From the PCA analysis shown in Fig. 5a, we can see that the downstream slope ($r=-0.66$, $p<0.001$) (Fig. 5c) and the relevant aspect ($r=-0.75$, $p<0.001$) of glaciers are the main factors responsible for the surface area losses, whereas elevation seems to have less influence. These findings agree with the few other studies investigating these relationships[19,35,36,37]. Whereas elevation is a proxy for temperature[35,37], aspect is a proxy for insolation[38], and slope is the key factor responsible for the gravitational driving stresses to which glaciers are subjected[3]. Lower slopes are supposed to induce lower glacier ice flow, thus allowing the development of stagnant ice conditions and consequently lower terminus retreat rates[3,39,40] and the development of supraglacial lakes[19,41]. In contrast, as we found here, under the climate warming pressure observed in the region for the last decades[8], higher slopes favor the shrinkage of glaciers because they are supposed to contribute to the higher glacier velocity[42].

Moreover, we observed that even glacier aspect is indirectly correlated with surface area losses, i.e., the largest shrinkages have been experienced mainly for E and W facing glaciers, and the shrinkages are decidedly smaller for S facing ones (Fig. 5a). Considering that S oriented glaciers are supposed to receive more solar radiation[38], the observed spatial shrinkage seems to be counterintuitive. However, we have to consider that, as their aspects deviates from S, the glaciers become significantly smaller (glacier aspect *vs* glacier size $r=-0.61$, $p< 0.001$) and steeper (glacier aspect *vs* glacier downstream slope $r=0.52$, $p< 0.01$) (all correlations are shown in Fig. SI4) because they are located in valleys perpendicular to the prevailing S-N direction of the monsoon. In our opinion, these valleys have been less hollowed, and the current glaciers are thus small and steep. Therefore, the observed correlation between aspect and glacier surface area losses indicates that gravitational stresses (conditioned by slopes) are higher for aspects far from S, and these, as observed above, are key factors in defining the rate of retreat of glaciers under the climatic pressure.

Although affected by a wide uncertainty, even at the scale of 20 survey lake basins we analyzed which morphometric factors were able to cause changes in surface areas. The correlation matrix among all of these variables is presented in Fig. SI4. We can see that the glacier slope ($r=-0.58$, $p<0.05$) and the relevant aspect ($r=-0.49$, $p<0.05$) of glaciers are the main factors responsible for the surface area losses, though, for the effect of larger uncertainty, these correlations are weaker than what we found at large scale.

Changes in snow cover duration. During the 2002-2012 period, we observed a decrease of the seasonal snow cover duration of -29% ($p<0.01$) within an elevation ranging from 4500 to 5500 m a.s.l. A similar behavior (even if we focus on the analysis exclusively) was found within the 20 survey lake basins (-27%, $p<0.01$). We found that the largest differences in snow cover duration occurred with W and S exposure and that the smallest occurred in the opposite orientations. Aspect ($r=0.23$, $p<0.001$) and elevation ($r=0.24$, $p<0.001$) are the main factors responsible for changes in snow cover duration, and we did not find any strong relationship with slope (all correlations are shown in Fig. SI5). In contrast to what we observed for glaciers, the snow cover duration at lower elevations decreased more due to higher temperatures that were recordable downstream, and the snow cover duration on S and W slopes decreased more than on N and E slopes, likely as a result of the higher solar irradiance received by these sides[43]. In the next paragraph changes observed in the seasonal snow cover duration are related to changes occurred in SO_4^{2-} and HCO_3^- concentrations of 20 survey lakes.

Changes in ionic concentrations. Once it is defined how variations recorded in glacier and snow cover duration have been conditioned by morphometric factors, we analyzed which of these factors are mainly responsible for changes observed in SO_4^{2-} and HCO_3^- concentrations in the survey lakes (Fig. 5b). Concerning the HCO_3^- concentrations, we did not find any relationship with the selected morphometric boundary conditions (Fig. 5b; Fig. SI6). Differently, Figure 5b reveals that glacier slope ($r=0.81$, $p<0.001$) (Fig. 5d) and relevant aspect ($r=0.66$, $p<0.01$) are well correlated with SO_4^{2-} concentration changes, whereas the direct relationship with the glacier surface areas losses is significant, but weaker ($r=-0.49$, $p<0.05$), probably affected by the high uncertainty connected with the measurements of small glaciers (all correlations are shown in Fig. SI6). From Figure SI6 we can also observe that the SO_4^{2-} concentration changes are not correlated with the basin slopes ($r=0.18$, $p>0.10$), as well as the direct relationship between glaciers slopes and basins slopes is weak ($r=0.44$, $p<0.05$) enforcing the causal link between the SO_4^{2-} variations and the glacier slopes. As indicated above, the glacier slope is also the main morphological feature able to explain the changes of the glacier surface area at higher and more representative scale (Fig. 5a and Fig. 5b). Therefore, we can assert that lake basins with steep glaciers oriented far from S have experienced higher glacier surface area losses and a concomitant SO_4^{2-} increases. Differently, the main decreases in snow cover duration occurred at lower elevations and on land with a S orientation, and they are less strictly dependent on the terrain slope (Fig. SI5 and Fig. SI6). Therefore, the changes observed in snow cover duration seem not to be the main cause of the observed SO_4^{2-} increases.

LCN9 experienced a particularly evident increase in ionic concentrations, which was even 4-fold in the case of SO_4^{2-} concentration (Fig. 3a). In this case, the reason for this huge change can also be found in that the LCN9 glacier has the most divergent aspect (SE-E, 110°) from the south orientation and has the highest glacier slope (36°) among the 20 survey lakes (Table1). Moreover, a synchronous temporal pattern of solutes and conductivity can be observed in LCN9

and LCN10 (Fig. 3a, 3b, 3c): the increases for both lakes should be driven by the same source, i.e., the glacier located within the LCN9 basin.

To further investigate the processes, which could have led to the SO_4^{2-} increase, we analyzed the associations between the main ions and how they changed during the last twenty years. We observed a highly significant positive relationship between SO_4^{2-} and $Ca^{2+} + Mg^{2+}$ in both in the early 1990s and in the early 2010s ($r = 0.85$ and 0.94, $p<0.001$) (Fig. SI3). On the contrary, the relationship between HCO_3^- and $Ca^{2+} + Mg^{2+}$ is weakly significant in the early 1990s ($r=0.39$, $p<0.05$) (Fig. SI3) and not significant in the early 2010s. The association between SO_4^{2-} and HCO_3^- was quite low in both periods ($r = 0.45$, $p=0.04$ and "not significant" in 2010s). The ratios of $(Ca^{2+} + Mg^{2+})/(Na^+ + K^+)$ was 7.2 in the early 1990s and 8.7 in the early 2010s, indicating the relative weakness of silicate dissolution. This is confirmed also by the lack of correlation between Si and $Na^+ + K^+$ in the 2010s ($r=0.17$). These results make evident an excess of SO_4^{2-} with respect to HCO_3^- in waters flowing in the drainage system of glaciers. This shift towards SO_4^{2-} enriched waters is even more evident in LCN9 and LCN10, as shown by the comparison of the ionic composition of lakes water in the 1990s and in the 2010s (Table SI1).

It is known that subglacial environments are dominated by carbonate dissolution and sulfide oxidation[44], often microbially mediated[45,46,47]. Through these processes, carbonate and sulfate concentrations in melt waters are greater than might be expected from the bedrock lithology.[48,49,50,51] A number of studies demonstrated that carbonation was mainly responsible of solute acquisition in the quick-flow component of the glacier hydrological system, while the enrichment in SO_4^{2-} seems to be strictly associated to delayed flow waters.[52] For these reasons, sulfate is considered an indicator of distributed type drainage beneath glaciers[53] and its increase, in most glacial environments, is an effect of increasing residence times and rock/water interaction[54]. In general, the oxidation of sulfide minerals and the dissolution of carbonates are partially coupled, since the hydrogen ions, originated from the sulfide oxidations, fuel the carbonate hydrolysis[53]. Our results, showing the lack of any relationship between SO_4^{2-} and HCO_3^-, suggest that the two processes are uncoupled in our system i.e. protons generated by sulfide oxidation do not dissolve carbonates. Furthermore, for most lakes the c-ratio was at values lower than 0.5 in 2010s and, particularly for LCN9 and LCN10, reached 0.2 in some years. These results suggest that other processes, likely in addition to carbonate dissolution and sulfide oxidation, occurred in the selected lake basins.

A process consistent with the elevated SO_4^{2-}, Ca^{2+} and Mg^{2+} concentrations found recently in lake waters is the pyrite oxidation coupled to the preferential weathering of calcium-silicate minerals which presence is documented in the study area. As suggested by Williams et al.[7] the Si produced from the weathering of calcium-silicate would be precipitated due to the effect of low temperature and this could explain the lack of correlation between SO_4^{2-} and Si, and the slight Si increase observed in the studied period.

304 The deglaciation observed in the study area has, first of all, increased the amount of meltwater causing a rising of
305 conductivity of lake waters. The disproportional increase of SO_4^{2-} compared to HCO_3^- may be explained by i) a change
306 in flow-paths and residence time of the meltwaters, leading to more favorable conditions for the oxidation of sulfides,
307 and ii) the pyrite oxidation followed by the weathering of calcium-silicate. We cannot exclude a role of microbial
308 populations at glacier beds if the conditions had become anoxic.[45,46,47] Figure SI7 shows the significant correlation
309 between H^+ differences and SO_4^{2-} differences ($r=0.72$, $p<0.001$) observed for the studied lakes, which can be interpreted
310 as further evidence of the importance of sulfide oxidation connected with the glacier retreat. At this regards it is
311 interesting to observe that Andersen[55] wrote that sulfide oxidation contributes to solute fluxes from glaciers to an extent
312 greater than might be expected from mineral abundance.

313 **Weathering and precipitation**.

314 Regarding the weathering occurring on the non glaciated part of the basin (periglacial environment), we analyzed
315 the behavior of the lakes (three) without glaciers within their basins. They showed a slight increase in SO_4^{2-}
316 concentration (+17% as median) (Fig. 2b), whereas higher increases were observed for lakes with glaciers within their
317 basins (+78% as median). These differences between the two groups were significant ($p<0.01$). The slight increase in
318 SO_4^{2-} concentration of lakes without glaciers could be attributed to the intensification of weathering rates caused by the
319 reduction of the snow cover duration[56], although we already discussed that this process is not the main driver of change.
320 Particularly, during the warmer season, on the sunny slopes, previously snow covered, an increase in soil temperature
321 from 0°C to 25°C could be expected, which could intensify the weathering rates by an order of magnitude.[57]
322 Weathering could be further enhanced by wetter reactive surface areas of minerals[58], but the decreasing precipitation
323 trend, as observed above over the last twenty years, does not support this possibility.

324 Nonetheless, the decreasing precipitation trend could have influenced the solute concentration in a different way:
325 lower but more concentrated runoff, could have slightly enriched the lakes[59]. These solute variations were caused by
326 enhanced weathering rates, or more probably by more concentrated runoff. Another mechanism considers that the
327 possible dropping of water-table depth, due to decreased groundwater recharge, resulting in more rock exposed to air
328 and enhanced mineral oxidation.[60] However they can be considered background increases that are common to all survey
329 lakes with respect to the higher increases observed in conjunction with glacier shrinkages.

330 **Annual chemical trends for two selected lakes.** Sulfate was the ion showing the most pronounced change in LCN9
331 and LCN10 (Fig. SI1, Table SI1). Hence we tested the possible dependence of the SO_4^{2-} concentrations in LCN9 on
332 monthly mean temperature, precipitation, and glacier melting, estimated by applying the T-index model[26]. We found
333 that SO_4^{2-} concentrations are only significantly correlated with the mean temperature in April ($r=0.74$, $p<0.001$). The

temperature and precipitation of the other months did not show any significant influence on the observed SO_4^{2-} concentrations (Fig. SI8). As for estimated glacier melt, we found that the only month that was significantly correlated, even in this case, is April ($r=0.74$, $p<0.001$) (Fig. 6a, 6b), whereas March shows a weak correlation ($r=0.45$, $p<0.1$) (Fig. 6a).

As shown above, significant increases in mean air temperature have been experienced over the last twenty years for the months of April, November and December.[9] However, April is the only month in which the increase has been effective, i.e., it has *de facto* favored the melting process. More precisely, the 0 °C isotherm in April shifted from around the mean elevation of glacier fronts to the mean elevation of glaciers located in the survey lake basins (Fig. 1b). In contrast, during the winter months, the temperature is so low that the 0 °C isotherm upward cannot have reached these elevations, and it cannot have had an effective impact on the melting processes. As a result, the T-index model estimates a significant increasing trend only for April ($p<0.01$). At this regards, the two main peaks of conductivity and major ion concentrations in 1998 and 2010 observed in Figure 3 are caused by the high temperature registered for the month of April of those years with subsequent high melting processes as estimated by the T-index model.

Previously, Lami et al.[5] observed a significant relationship between the enhanced conductivity detected in LCN9 and LCN10 and the annual temperature until 2008. Here we confirm the observations of these authors until 2013 considering the temperature as the main climatic driver of change. Besides that, we found the temperature of April is the effective drive of the observed enhanced glacier melting process, which is considered here the main factor responsible for the observed increase of sulfate concentrations.

The chemical variations observed in lake water in the last two decades represent a response of these fragile ecosystems to climate change. In particular, glacier retreat proved to be the main driver of the solute increase affecting the lakes. Major ion concentrations, and sulfate at a higher extent, significantly rose in lake water. Even if these changes do not pose a direct and immediate threat to the biota, they occurred in a limited time span, and significantly modified the average chemical composition of lake water. For these reasons, the lakes and the main factors driving their variability should be regularly monitored in the future, also in relation to the lake role as ecosystem services.

SUPPORTING INFORMATION

A table showing the chemical and topographic features of 20 survey lakes. A figure showing the relative contribution of various ions to the total ionic content of LCN 9 in the early 1990s and in the early 2010s. ALOS 2008 imagery of LCN68 and LCN67 basins. Correlation matrixes among chemical species, land covers, and topographic boundary conditions. A figure showing the relationship between the $SO4^{2-}$ and the H^+ concentration changes observed in all

363 survey lakes between 1990s and 2010s. A figure showing the PCAs among $SO4^{2-}$ and HCO_3^- concentrations of LCN9,
364 mean temperature, and precipitation for each month of the year.

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TABLES

TABLE 1. Mean morphometric features and land cover for the 20 selected lakes (reference year 1992). For LCN 10 data are referred to the sub-basin draining directly into the lake (see graphical abstract).

Morphometry	**20 survey lakes Median (range)**	**LCN9**	**LCN10**
Lake elevation (m a.s.l.)	5010(4466-5427)	5209	5058
Lake area (km^2)	0.02(0.003-0.581)	0.005	0.013
Basin area (km^2)	0.8(0.1-41)	0.66	0.32
Basin slope (°)	29(9-35)	35	25
Basin aspect (°)	160(120-231)	120	120
Basin mean elevation (m a.s.l.)	5204(4843-5539)	5403	5102
Glacier area (km^2)	0.31(0.03-6.4)	0.11	-
Glacier slope (°)	26(15-36)	36	-
Glacier aspect (°)	151(110-221)	110	-
Glacier mean elevation (m a.s.l.)	5326(5087-5581)	5463	-
Land cover	**20 survey lakes Mean (range)**	**LCN9**	**LCN10**
Glacier (%)	18(0-50)	17	0
Bare rock (%)	28(0-66)	30	25
Bare soil (%)	40(9-74)	53	75
Shrub vegetation (%)	14(0-63)	0	0

TABLE 2. Median values of conductivity, selected ionic concentrations, and c-ratio (see the text for the explanation) of the survey lakes sampled during the early 1990s and during the early 2010s. In brackets the standard deviation.

Feature	Early 1990s	Early 2010s	Difference (%)
Conductivity (µS cm^{-1}, 20 °C)	24.7 (16)	36.4 (27)	+47 ($p<0.001$)
SO42- (µeq L^{-1})	80 (129)	126 (245)	+57 ($p<0.001$)
HCO3- (µeq L^{-1})	128 (54)	141 (61)	+11 ($p<0.001$)
Ca2++Mg2+ (µeq L^{-1})	221 (124)	331 (224)	+41 ($p<0.001$)
H+ (µeq L^{-1})	0.03 (0.02)	0.07 (0.02)	+ 126% ($p<0.01$)
c-ratio	0.66 (0.22)	0.55 (0.26)	-18 ($p<0.01$)

FIGURE

FIGURE 1. a) Location of the study area in the Himalayas. b) Hypsometric curve of SNP and altitudinal glacier distribution. Along this curve, the location of 20 selected lakes is shown. The 0 °C isotherms corresponding to the mean monthly temperature in April are plotted for the years 1994 and 2013 according to the observed T trends and lapse rates[9]. c) Focused map on the spatial distribution of lakes and glaciers in Sagarmatha National Park (SNP) and the location of the Pyramid meteorological station.

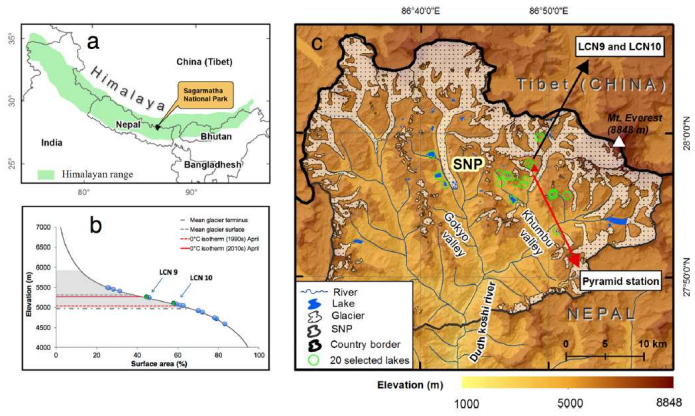


FIGURE 2. Scatter-plots between a) conductivity, b) SO_4^{2-}, c) HCO_3^-, d) Ca^{2+}+Mg^{2+}, e) H^+, and f) c-ratio of the survey lakes sampled during the early 1990s and the early 2010s. The orange points represent lakes without glaciers within the basin.

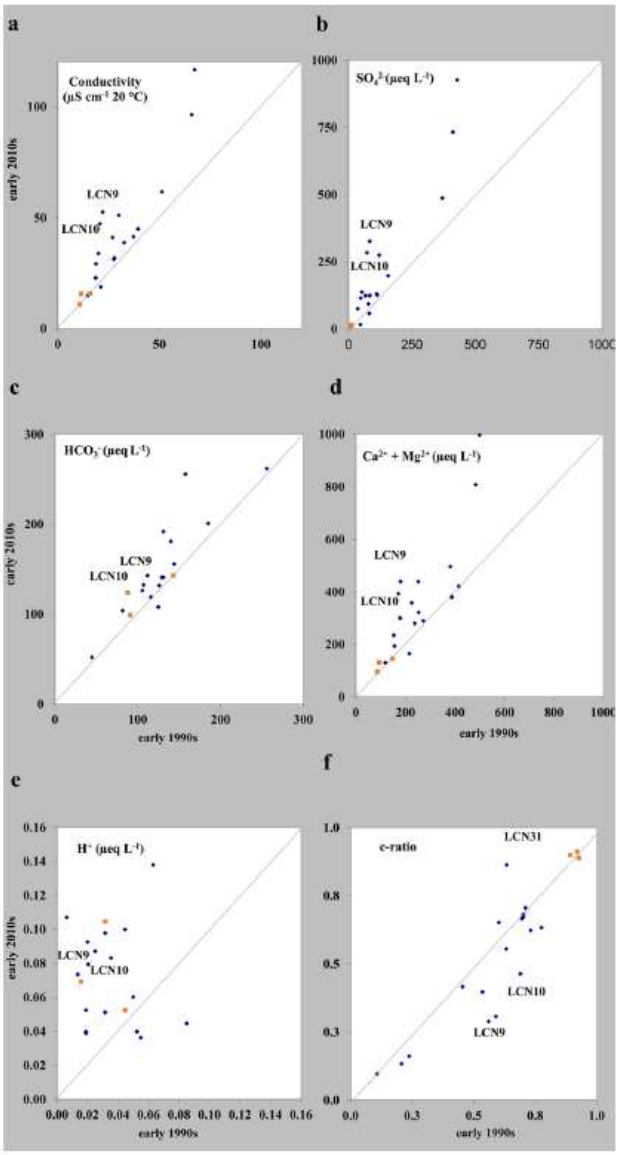


FIGURE 3. Long-term trends of the annual values of a) conductivity, b) SO_4^{2-}, c) HCO_3^- d) $Ca^{2+}+Mg^{2+}$, e) H^+ concentrations, and f) c-ratio in LCN9 (red) and LCN10 (blue).

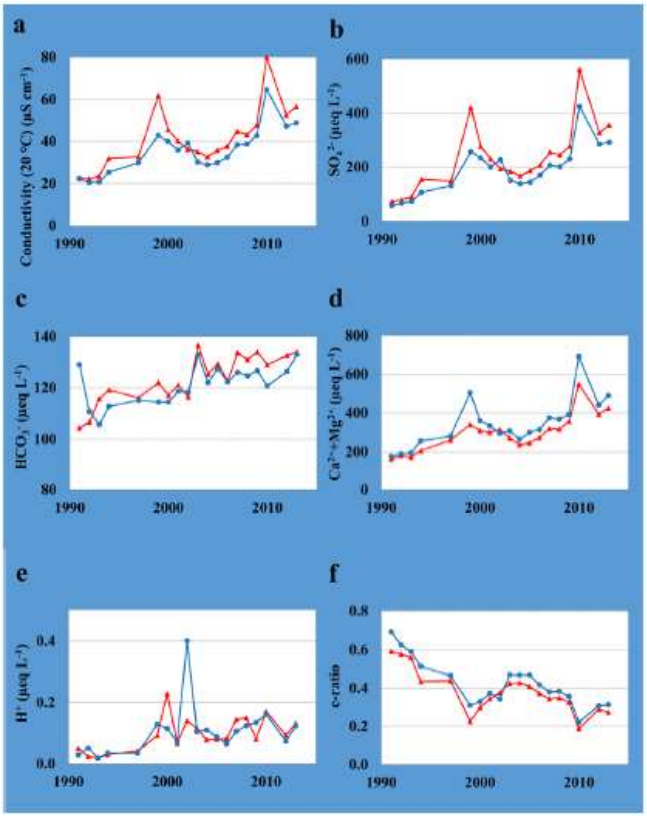
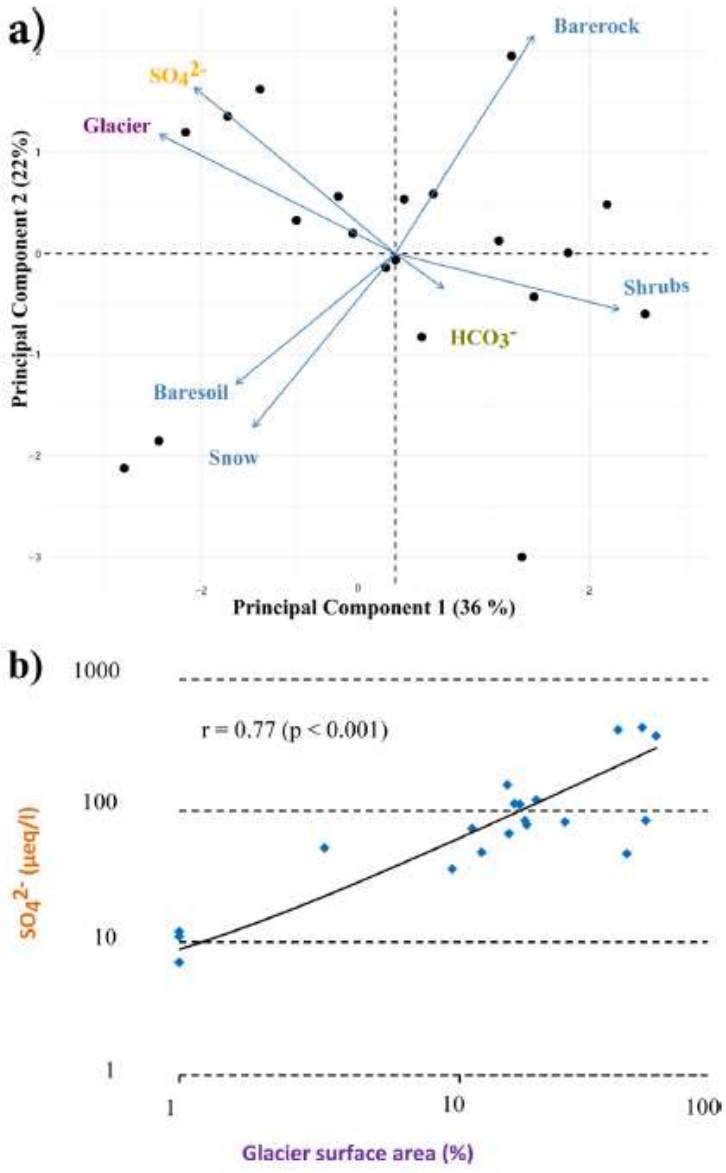


FIGURE 4. a) PCA among SO_4^{2-}, HCO_3^- concentrations of the survey lakes (early 1990s), land cover, and snow cover duration (*Glacier*: glacier coverage during the early1990s; *Baresoil, Barerock, Shrubs*: coverage for 2008; *Snow*: mean snow cover duration for each basin during the 2010s). b) The most significant correlation that emerged from the above PCA.



555 FIGURE 5. PCAs among morphometric parameters and a) glacier surface area variations (Δ*Glacier)* that occurred in
556 the last twenty years in the overall SNP (data from Thakuri et al.[10]); b) relative SO_4^{2-} and HCO_3^- variations (Δ SO_4^{2-},
557 ΔHCO_3^-) that occurred in the same period for the 20 survey lakes; (*Glacier_size*=size of glaciers; *Elev_max,*
558 *Elev_mean, Elev_min*: maximum, mean, and minimum glacier elevation, respectively; *Slope*, *Slope_down, Slope_up*:
559 mean, downstream, and upstream glacier slope, respectively; *Aspect*: mean glacier aspect). The lower graphs (c and d)
560 show the most significant correlations that emerged from the relevant PCAs shown above.

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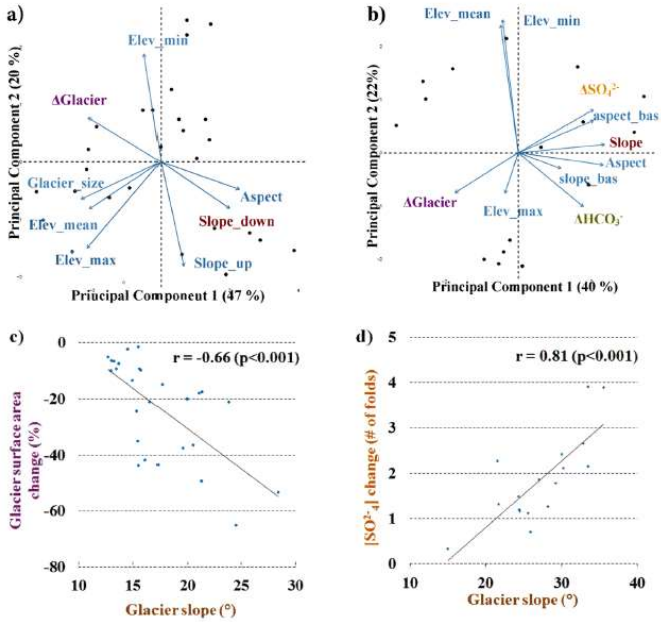

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FIGURE 6. a) PCA among $SO4^{2-}$ and HCO_3^- concentrations of LCN9 sampled annually in late September or October (*LCN9_ $SO4^{2-}$*, *LCN9_* HCO_3^-, respectively) and the cumulated glacier melt ($melt_i$) modeled for each month (*i*) of the year. b) The lower graph shows the most significant correlation that emerged from the PCA: *$SO4^{2-}$_LCN9* vs *melt4*.